

Aerosol Extinction:

Size, Relative Humidity, Composition, and Heterogeneous Chemistry

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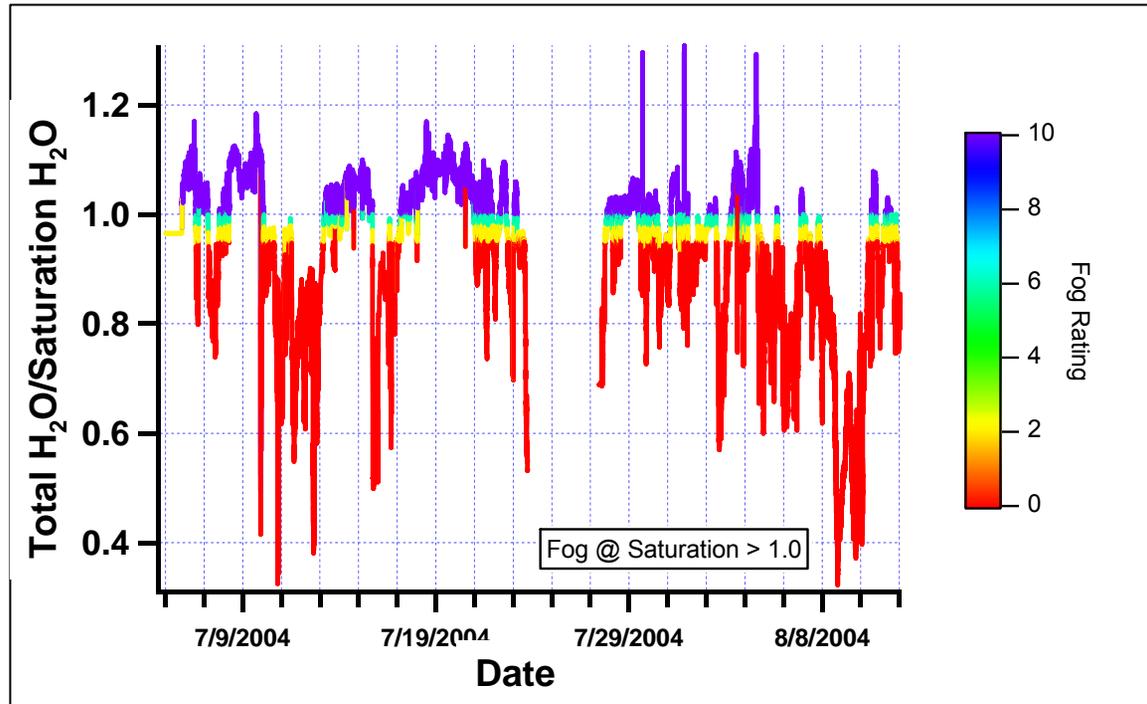
Boulder, CO

RV Ronald H Brown Data Workshop

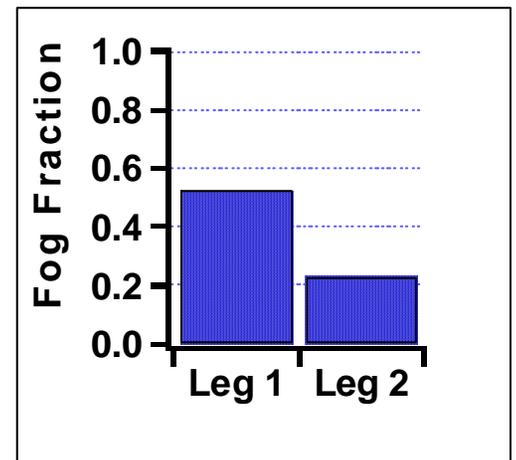
NEAQS-ITCT 2004

March 8th 2005

Fog



- A. $RH(T)$ (Internal CRD measurement) \rightarrow Total H₂O
- B. T (ambient) \rightarrow Saturation H₂O (vapor pressure)
- C. Total H₂O – Saturation H₂O \rightarrow LWC
- D. $(\text{Total H}_2\text{O} - \text{LWC})/\text{Saturation H}_2\text{O} \rightarrow$ Ambient RH
- E. $(\text{LWC}/\text{m}^3, \#/\text{m}^3 \text{ fog droplets}) \rightarrow$ Surface Area



Projects

- 1. Aerosol Optical Properties Closure**
- 2. Radiative Forcing**
 - Relative Humidity and Composition**
 - AOD**
- 3. Fog/Cloud Chemistry**
- 4. Plume Processing**
 - Fog Interface (e.g. SO_2/SO_4 and VOC)**
 - Photochemical Age**

Motivation (Cavity Ring-down Aerosol Spectrometer)

☆ Direct Measurement of Aerosol Extinction

$$\sigma_{ep} = \sigma_{ap} + \sigma_{sp} \quad AOD = \int \sigma_{ep}(z) dz \quad \omega = \frac{\sigma_{sp}}{\sigma_{ep}}$$

Extinction Aerosol Optical Depth single scattering albedo

Forcing

Direct effect

$$\Delta_a F \uparrow = \frac{1}{2} F_T T^2 (1 - A_c) \left[\omega_{RH} \beta_a (1 - R_s)^2 - (1 - \omega_{RH}) R_s \right] AOD_{RH}$$

Clouds (ΔA_c = indirect and semi-direct effect)

Key Point: Extinction and RH dependence everywhere!

Measured Parameters (Cavity Ring-down Aerosol Spectrometer)

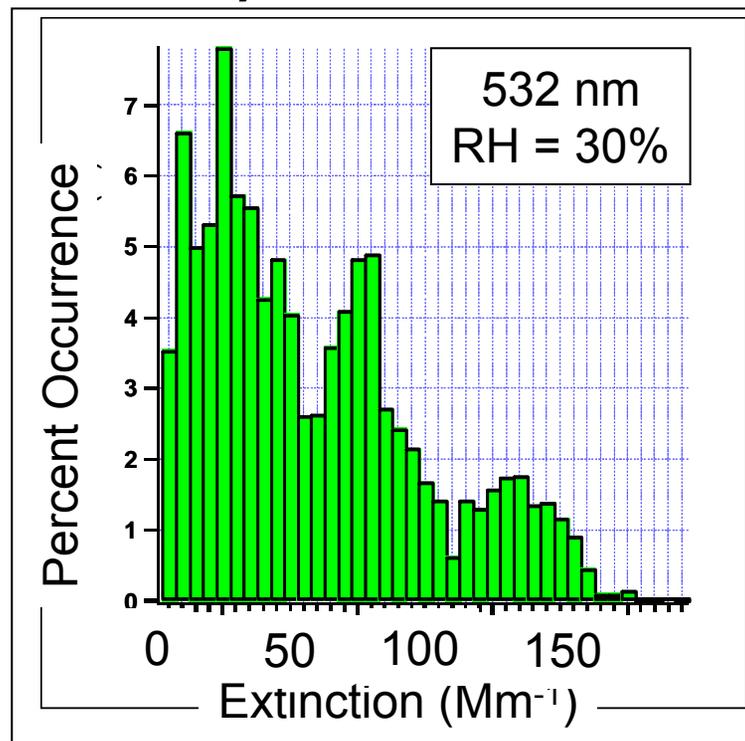
1. σ_{ep} : extinction coefficient
355nm, 532nm, 683nm, 1064nm

2. α_{ep} : wavelength dependence

$$\alpha_{ext} = - \frac{\log\left(\frac{\sigma_{ep}(1)}{\sigma_{ep}(2)}\right)}{\log\left(\frac{\lambda(1)}{\lambda(2)}\right)}$$

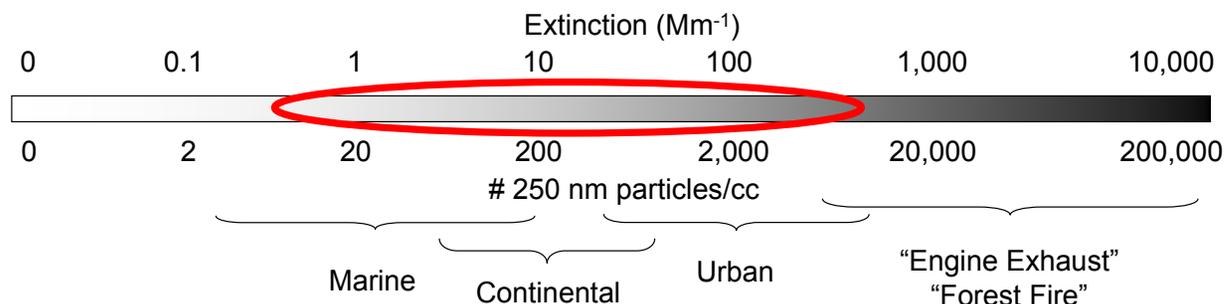
3. $f(RH)$: Relative Humidity Dependence

$$fRH_{ext} = \frac{\sigma_{ep}(Humidified)}{\sigma_{ep}(Dry)}$$



Units (extinction coefficient)

$$1Mm^{-1} = 10^{-3} km^{-1} = 10^{-8} cm^{-1}$$



Parameters (Cavity Ring-down Aerosol Spectrometer)

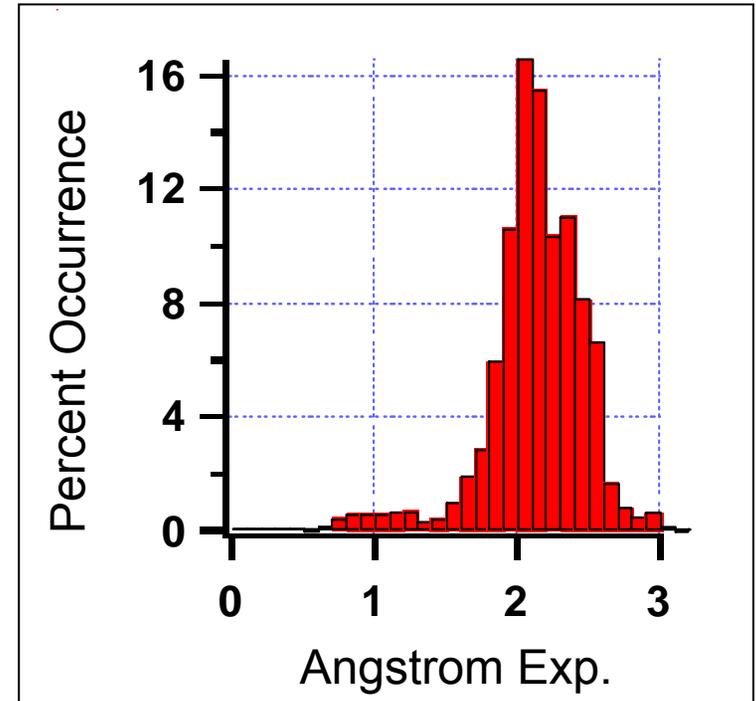
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$$\begin{array}{l} 355\text{nm} \\ 532\text{nm} \\ 683\text{nm} \\ 1064\text{nm} \end{array} \rightarrow \alpha_{ext} = -\frac{\log\left(\frac{\sigma_{ep}(1)}{\sigma_{ep}(2)}\right)}{\log\left(\frac{\lambda(1)}{\lambda(2)}\right)}$$

3. $f(RH)$: Relative Humidity Dependence

$$fRH_{ext} = \frac{\sigma_{ep}(\text{Humidified})}{\sigma_{ep}(\text{Dry})}$$



Angstrom Exponent (Wavelength Dependence)

1. Angstrom Exponent decreases with particle size
2. Low Angstrom Exponent more efficient interaction with full solar spectrum

Parameters (Cavity Ring-down Aerosol Spectrometer)

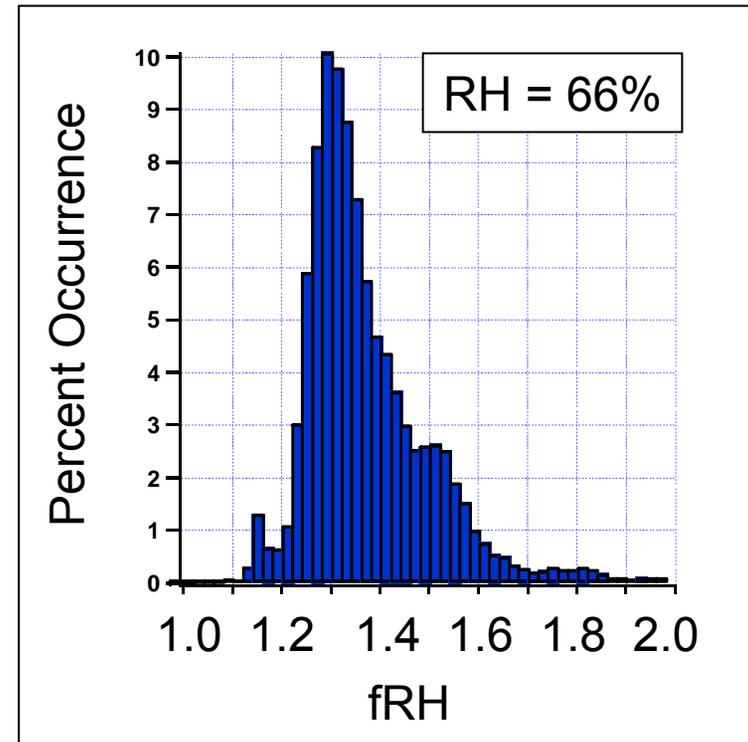
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Complimentary Measurements

σ_{sp} : scattering coefficient
@ 450nm, 550nm, 700nm

σ_{ap} : absorption coefficient
@ 467nm, 530nm, 660nm

Size distribution

Aerosol Composition (AMS, PILS, OCEC)

PMEL/UW/UI

PMEL/UW

PMEL/UW/AL

PMEL

1. Aerosol Optical Properties Closure (532 nm/550 nm)

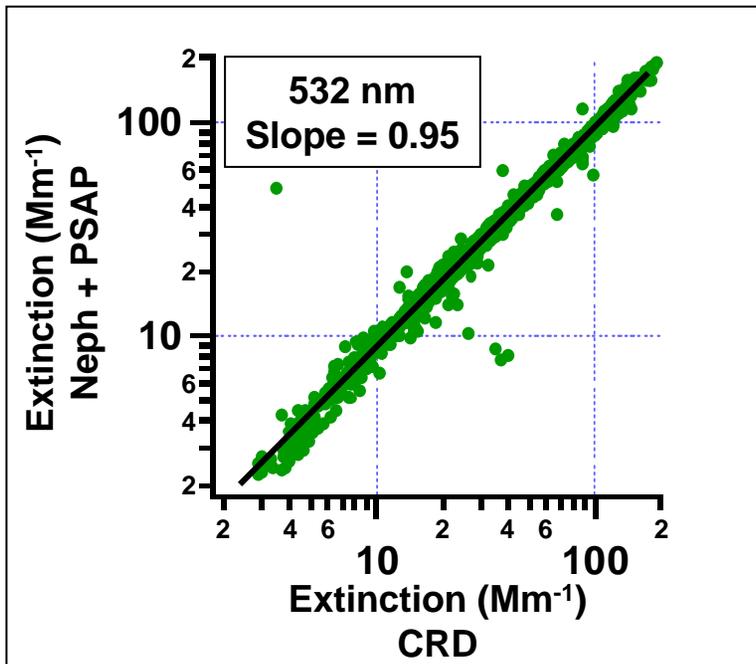
$$\sigma_{ep}(\text{CRD}) = \sigma_{sp}(\text{Neph}) + \sigma_{ap}(\text{PSAP})$$

$$\text{Uncertainty } \sigma_{(ap+sp)} = 7\%$$

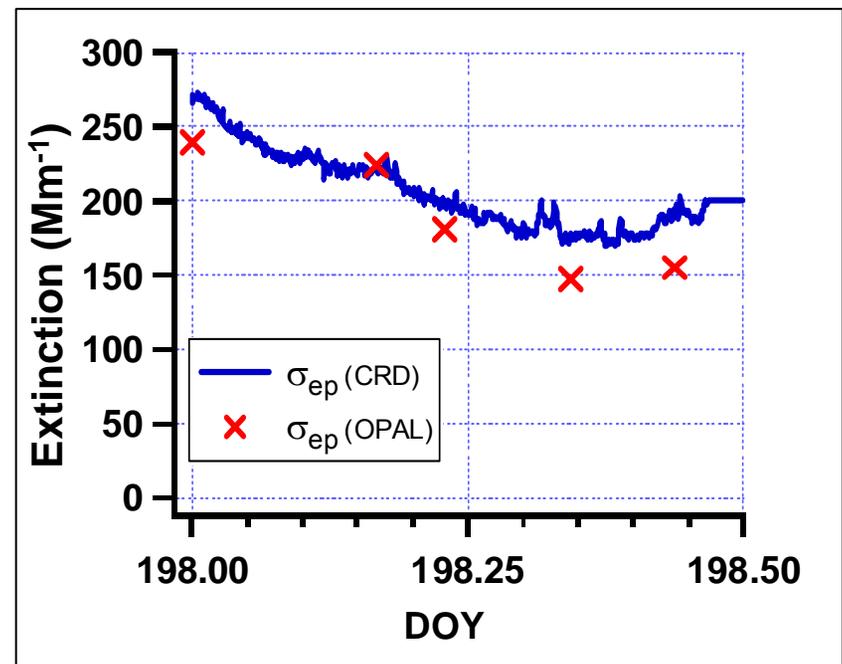
Difference in forcing $\approx +0.25 \text{ W/m}^2$ (Ocean)

$\approx +0.28 \text{ W/m}^2$ (Forest)

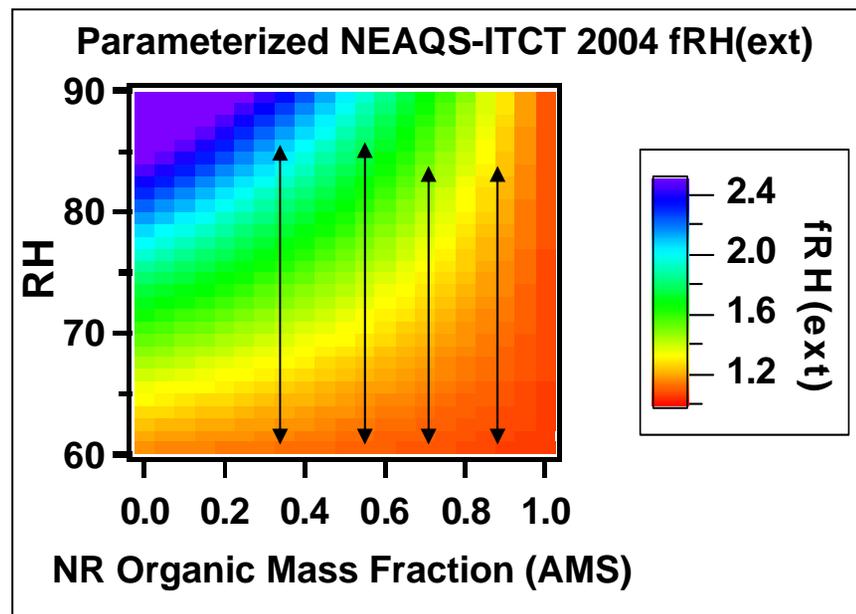
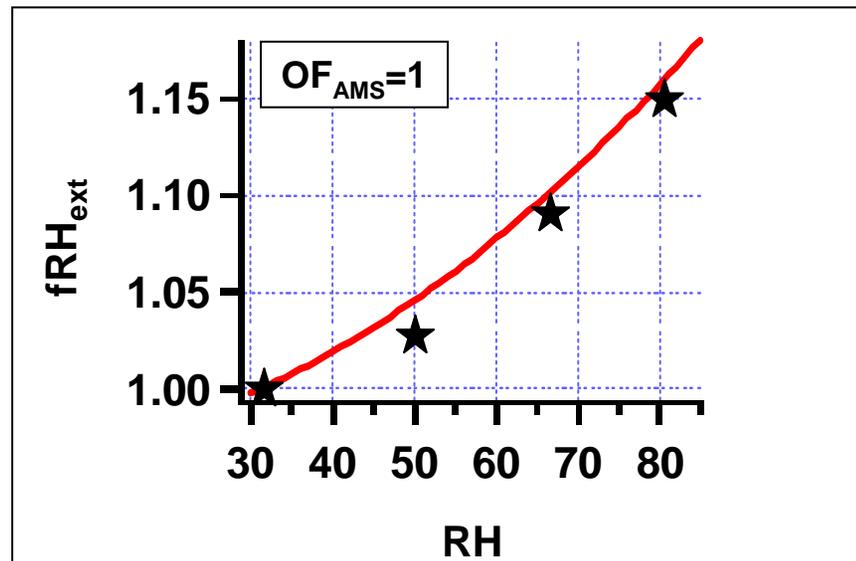
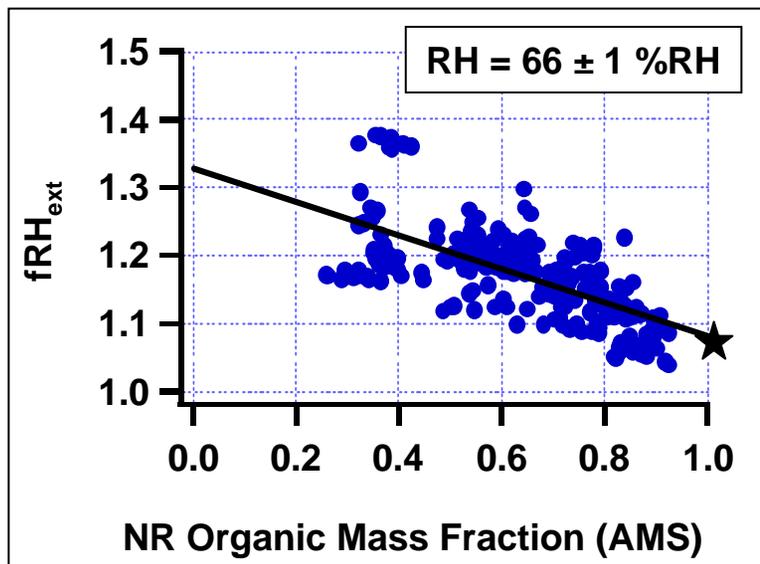
In-Situ Comparison



Remote Sensing Comparison



2. fRH (Relative Humidity Dependence)



☆ Observed composition dependence

Growth Factor (RH)

Mie Scattering

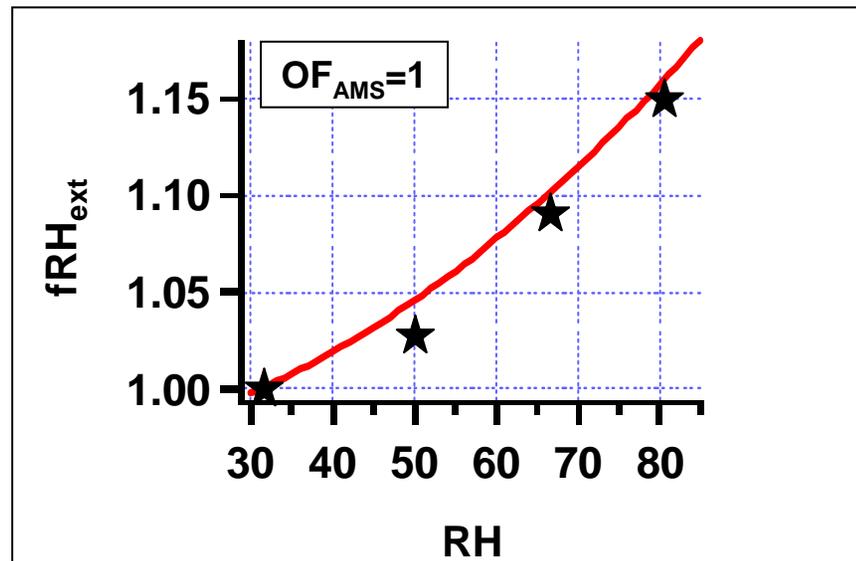
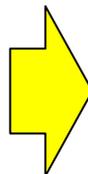
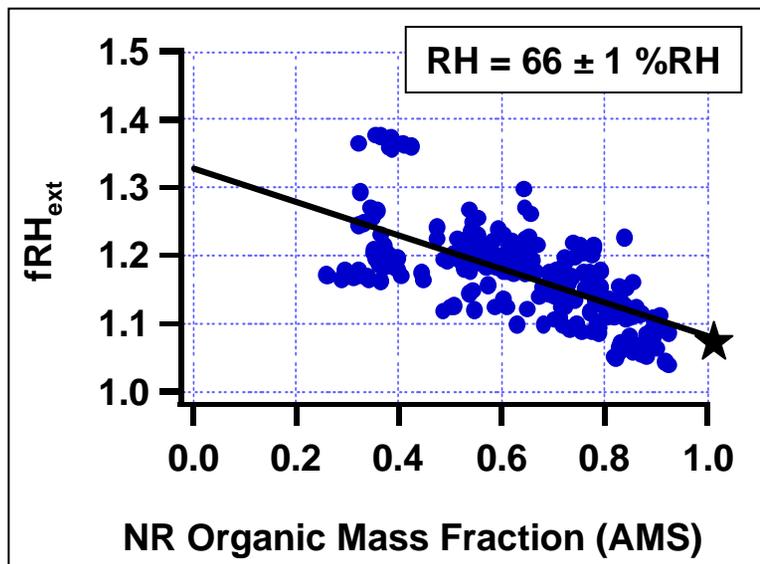
Refractive Index

Non-organic composition

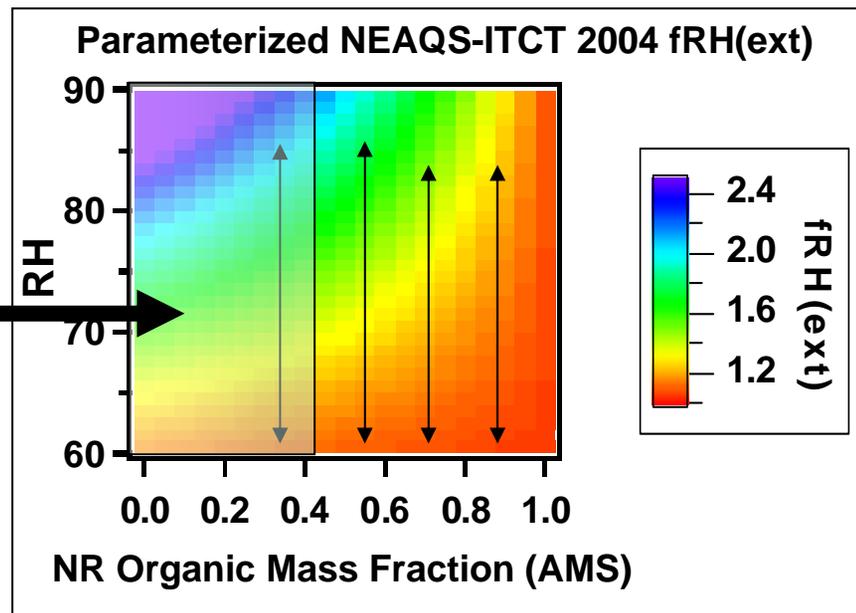
Organic composition

Size Distribution

2. fRH (Relative Humidity Dependence)

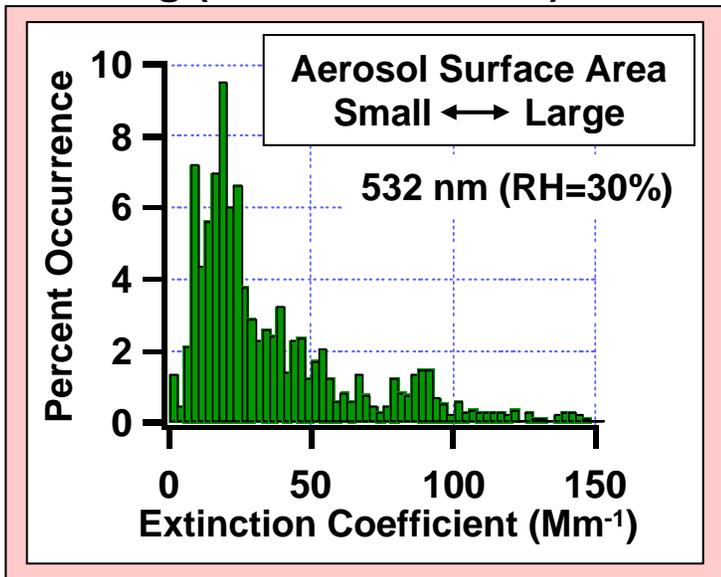


limitations



3. Fog/Cloud Chemistry (σ_{ep})

Non-Fog (Saturation < 0.95)

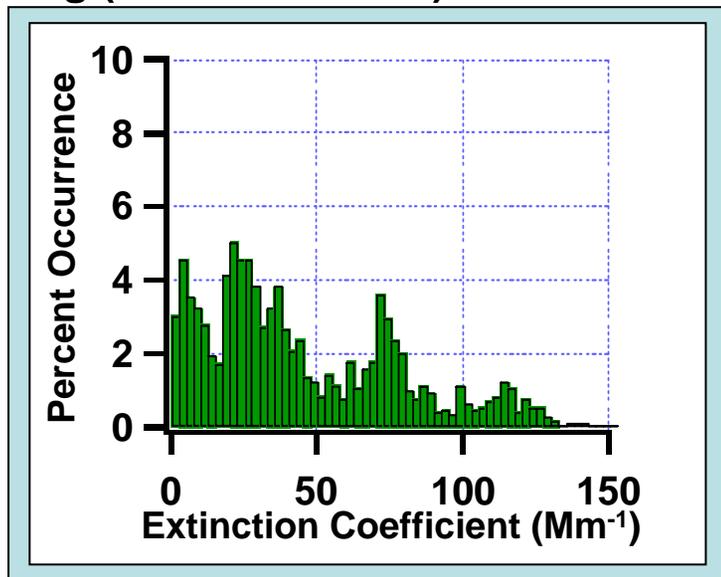


A. Example ($n=1.4$)

Increase in aerosol mass loading

$$\left. \begin{array}{l} \sigma_{ep} \ 85 \ Mm^{-1} \approx 11.5 \ \mu g/m^3 \\ \sigma_{ep} \ 117 \ Mm^{-1} \approx 15.3 \ \mu g/m^3 \end{array} \right\} \Delta M = 30\%$$

Fog (Saturation > 1.0)



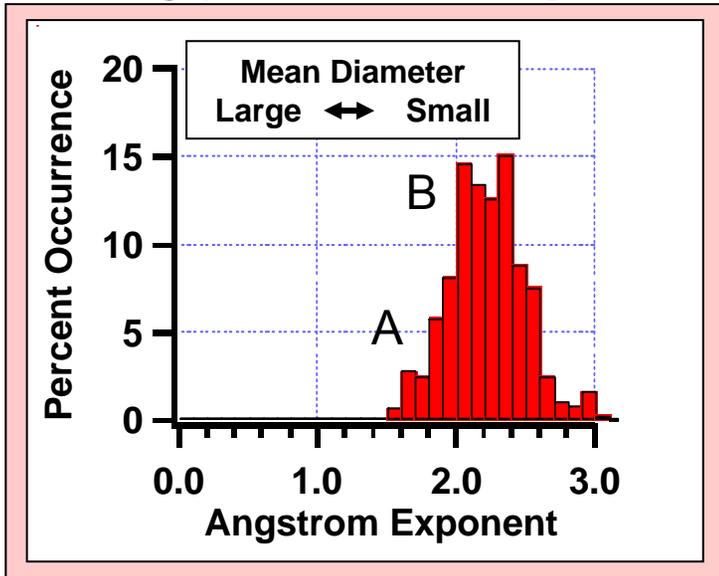
B. Mean

	Mean Extinction	Est. Mass $\mu g/m^3$	Δ Mass
Non- Fog	30 (Mm^{-1})	3.9	n/a
Fog	35 (Mm^{-1})	4.6	18%

Calculation: log-normal distribution (width = $0.25 \ \mu m$)
Mean diameter fixed (angstrom exponent = 2.1)

3. Fog/Cloud Chemistry (α)

Non-Fog (Saturation < 0.95)



A. Large Particles (Fog)

July 14th (DOY 196), Extinction $\approx 80 \text{ Mm}^{-1}$
 July 21st (DOY 203), Extinction $\approx 110 \text{ Mm}^{-1}$
 July 30th (DOY 212), Extinction $\approx 120 \text{ Mm}^{-1}$
 Aug 10th (DOY 223), Extinction $\approx 20 \text{ Mm}^{-1}$

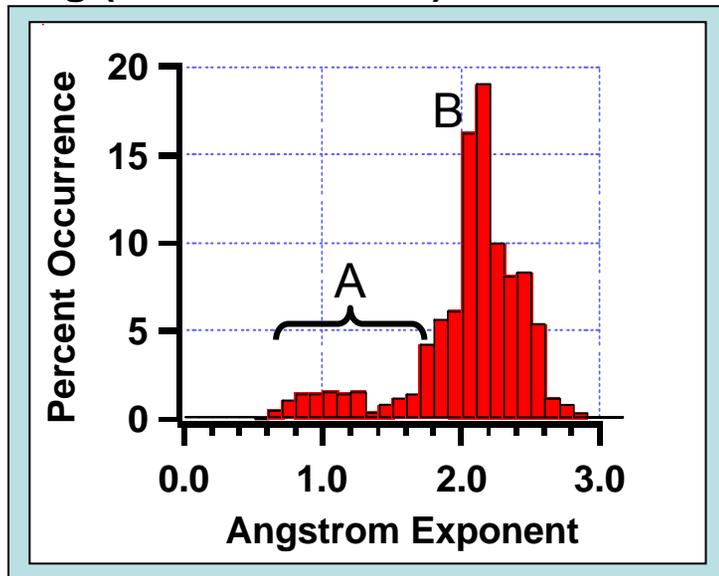
Shift to larger particles

Example ($n=1.4$)

Mean optical diameter (2.0) = $0.37 \mu\text{m}$

Mean optical diameter (1.0) = $0.53 \mu\text{m}$

Fog (Saturation > 1.0)



B. Fog Processing (Larger particles)

	Mean α	Mean Particle Diameter	Δ Volume
Non- Fog	2.25	$0.19 \mu\text{m}$	n/a
Fog	2.15	$0.21 \mu\text{m}$	25%

Calculation: log-normal distribution (width = $0.25 \mu\text{m}$)
 Number of particles fixed

3. Fog/Cloud Chemistry (fRH)

$\lambda = 532$ nm (main and reference)

Filtering:

Main RH: $66 \pm 1\%$ RH

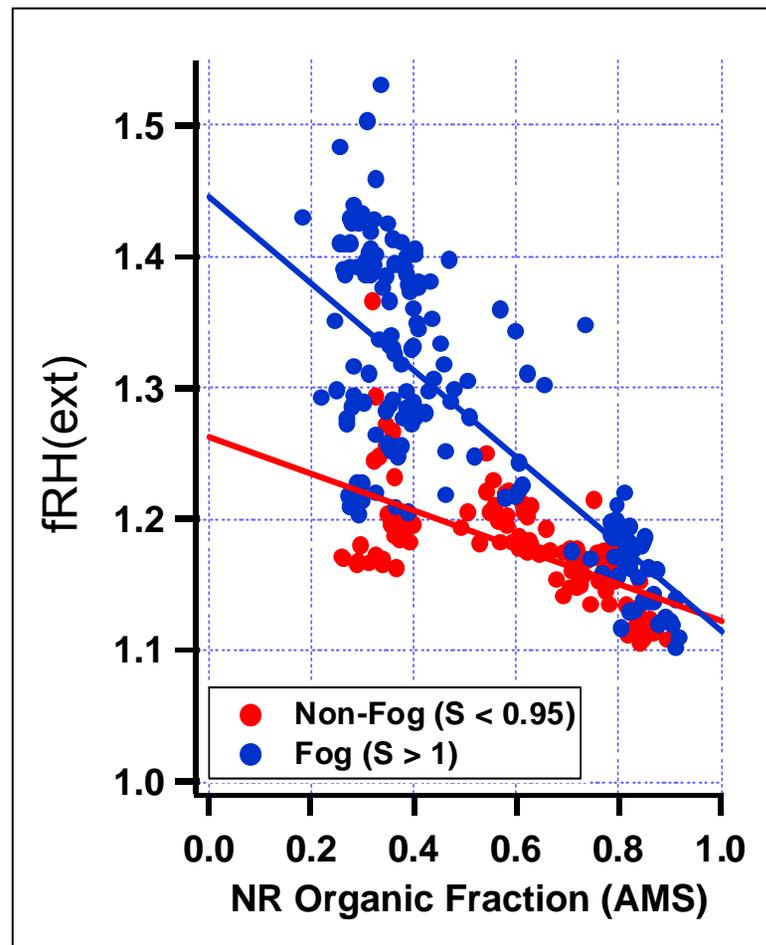
Angstrom Exponent: 2.2 to 2.4

Saturation: $S < 0.95$ and $S > 1$

Significant variation in fRH/OF
 especially at $0.2 < OF < 0.4$
 Gas phase chemistry
 Aerosol composition

Organic Fraction	fRH @ 66% RH		fRH* @ 85% RH	
	Non- Fog	Fog	Non- Fog	Fog
0	1.27	1.45	2.2	2.8
0.5	1.20	1.26	1.8	2.1
1	1.12	1.12	1.18	1.18

*estimate from OF = 0.3, 0.5, 0.7, 0.9 RH dependence



Key Points

1. Further reduction in uncertainty of in-situ aerosol optical properties is required.
 2. fRH (RH dependence) measured
 - a. Time resolution = 1 min
 - b. Sensitivity = 3%
 3. Relative humidity dependence of extinction (and scattering) on aerosol composition provides new and detailed information.
 - a. Speciated RH dependence from atmospheric measurements (e.g. Organics, Sulfate (?))
 4. Heterogeneous chemistry has a significant effect on aerosol optical properties, RH dependence, composition, and size distribution.
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Application

1. Link atmospheric gas phase chemistry to aerosol chemical and optical properties
2. Speciation of aerosol forcing including RH dependence
 - Sulfate: improved with proper separation of components
 - Organics: add appropriate RH dependence for organics
 - Heterogeneous chemistry: different composition and RH dependence

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